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LETTER TO THE EDITOR

Change in dielectric constant along and perpendicular to induced strain in a supercooled liquid and in amorphous media

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Abstract. Brillouin spectroscopy in amorphous media for different angles offers a direct way to determine the difference between changes in dielectric constant along and perpendicular to induced strain. Here the method is applied to quartz glass and supercooled liquid o-terphenyl. It is found for liquid o-terphenyl that the change in dielectric constant along the induced displacement is smaller than the change in dielectric constant perpendicular to the induced displacement, as it is also in quartz.

The elasto-optic coupling coefficients are of considerable interest basically, and for applications. The change in dielectric properties ϵ_{ij} of an amorphous medium upon strain is given by [1-3]

$$\delta\epsilon_{ij} = -\epsilon^2 \left[2P_{44}U_{ij} + P_{12} \left(\sum_k U_{kk} \right) \delta_{ij} \right].$$

Here U_{ij} are the components of the strain tensor defined by the spatial derivatives of the local displacement vector u [2]

$$U_{ij} = \frac{1}{2} \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right).$$

The elasto-optic coefficients P (*Pockels coefficients*) which describe the change of ϵ_{ij} with induced strain are for isotropic media: P_{12} for changes of the components of ϵ_{ij} perpendicular to the components of U_{ij} , P_{11} for the parallel component, and P_{44} for the off-diagonal change induced by shear; due to the Cauchy condition $P_{44} = \frac{1}{2}(P_{11} - P_{12})$. By applying static displacement [4] it is possible to measure the difference between P_{11} and P_{12} or sign of P_{44} for solid amorphous media. Mueller [3] has studied a large variety of oxide glasses, mostly based on SiO_2 .

The sign of P_{44} is negative in most glasses, as it is in quartz, but some amorphous polymers have $P_{44} > 0$ [5,6]. To our knowledge, the sign of P_{44} is not known for

any liquid. Here we describe a way of determining this for amorphous substances in general by Brillouin spectroscopy, and we demonstrate that this method can be extended to liquids in the quasistatic region ($\omega\tau > 1$).

We first provide the theoretical background to the problem. The Brillouin scattering intensities I_{ij} from longitudinal and transverse acoustic phonons (L, T) are given by [1]

$$I_{HH}^L = I_0 V_S \pi^2 k_B T \frac{\epsilon^4}{\lambda_0^4 \rho c_L^2} [P_{44} + (P_{12} + P_{44}) \cos \theta]^2 \quad I_{HV}^L = 0 \quad (1)$$

$$I_{VH}^T = I_{HV}^T = I_0 V_S \pi^2 k_B T \frac{\epsilon^4}{\lambda_0^4 \rho c_T^2} \left[P_{44} \cos \left(\frac{\theta}{2} \right) \right]^2 \quad I_{VV}^T = I_{HH}^T = 0. \quad (2)$$

where V and H refer to vertical and horizontal planes of polarizations relative to the scattering plane. The variables are defined as follows: I_0 is the incident light intensity, V_S the scattering volume, $\epsilon = n^2$ the average dielectric constant of the material, λ_0 the wavelength of incident light in a vacuum, ρ the density, c_L the longitudinal and c_T the transverse sound velocities, and θ the internal scattering angle.

The frequency shifts $\delta\nu^{L,T}$ of the two Brillouin lines due to the two sound waves in the medium are given by [1] as

$$\delta\nu^{L,T} = c_{L,T} \frac{2\sqrt{\epsilon}}{\lambda_0} \sin \left(\frac{\theta}{2} \right) \quad (3)$$

with $c_L > c_T$. As an example, figure 1 shows Brillouin spectra for o-terphenyl.

If the polarization of the incoming beam is parallel to the scattering plane and if there is no polarization analysis in the measurement of intensity I , the ratio of longitudinal to transverse intensity from (1) and (2) is

$$\frac{I_{HO}^L}{I_{HO}^T} = \frac{I_{HH}^L}{I_{HV}^T} = \left[\frac{c_T P_{44} + (P_{12} + P_{44}) \cos \theta}{c_L P_{44} \cos(\theta/2)} \right]^2. \quad (4)$$

The last expression has a minimum, equal to zero, when the scattering angle θ ($0 \leq \theta \leq 180^\circ$) is defined by the equation

$$\cos \theta = - \left(1 + \frac{P_{12}}{P_{44}} \right)^{-1}. \quad (5)$$

This equation has two solutions for positive and negative P_{12}/P_{44} respectively. Since P_{12} is positive for most amorphous media and larger than P_{44} in the cases we know, the expression on the right-hand side of (5) has the opposite sign relative to P_{44} . If P_{44} is positive, the minimum is reached at a scattering angle larger than 90° , whereas the minimum is reached at an angle smaller than 90° for negative P_{44} . The graph of (4) for both positive and negative P_{44} is shown in figures 2(a) and 2(b). For the plane of polarization parallel to the scattering plane, equations (4) and (5) imply that the Brillouin intensity of the scattering from the longitudinal phonons becomes zero at a certain angle, defined by the elasto-optic properties of the medium. For quartz glass this angle is 70° .

Although shear strain cannot be sustained statically in a liquid because of the finite value of shear viscosity, TA sound waves remain propagating in the frequency domain

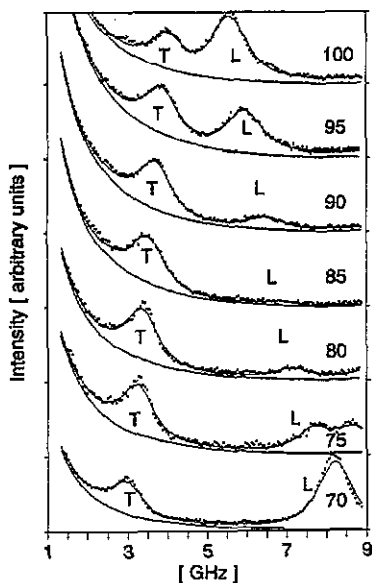


Figure 1. Brillouin spectra for supercooled o-terphenyl in HO polarization at room temperature for different scattering angles. L and T show the scattering from longitudinal and transverse sound waves respectively. The longitudinal sound wave appears in the other half of the free spectral range, which is 16.5 GHz. Points are experimental data, solid lines show the result of the fit and the fitted background.

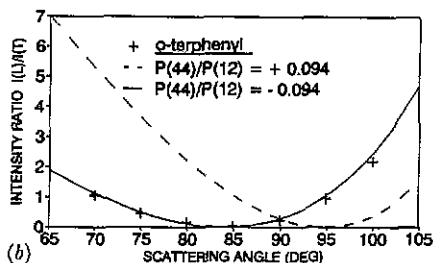
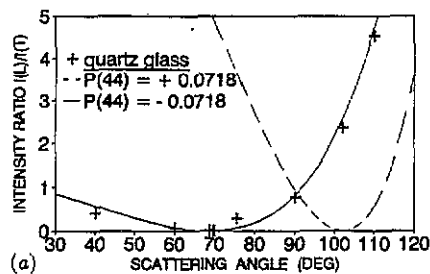


Figure 2. The ratio of longitudinal and transverse intensities measured and calculated from (4) for either sign of P_{44} for (a) quartz glass and (b) o-terphenyl.

$\omega\tau > 1$ [8]. For a glass-forming liquid like o-terphenyl, this region is accessible by Brillouin scattering at room temperature (relaxation time $\tau \approx 10^{-10}$ s [9]).

We now outline our experimental procedure and give our results. The experiment was first carried out on quartz glass (Suprasil). A cylindrical sample (diameter 3 cm) of optical quality was used. As a liquid suited for this study, an o-terphenyl sample was prepared by distilling the material (Aldrich) under vacuum into a cylindrical Pyrex tube of diameter 2.4 cm, and then sealing the tube. It then remains easily supercooled at room temperature. The experimental arrangement has been described by Loheider *et al* [10]; therefore we concentrate on a few important details of set-up and control:

The measurements of the longitudinal and transverse Brillouin lines were done at room temperature by using a single-mode ($\lambda = 514.5$ nm for quartz, 488 nm for o-terphenyl) Ar-ion laser (Spectra-Physics 164) and a piezoelectrically scanned five-pass (triple-pass for quartz) Fabry-Perot interferometer (Burleigh RC-170). Two spherical lenses of focal length 50 cm and 20 cm were used in the incident and scattered beams, respectively. Horizontal polarization of the incident beam was produced by a $\lambda/2$ plate and secured by a subsequent Nicol prism. The experiment was carried out under control of an Olivetti 380 XP1 (and an RT-11 for the quartz measurements).

For the quartz spectra the relative intensities I_{HO}^L/I_{HO}^T were calculated as integrals over the Brillouin lines minus noise level. Brillouin spectra of o-terphenyl are shown

in figure 1. The o-terphenyl spectra were fitted individually using the same program as described earlier by Soltwisch *et al* [11], assuming broadened Lorentzians plus background for the LA and TA lines. The same shape was used to describe the quasielastic line; this line contributes a background at the position of the TA line which starts to rise steeply towards lower frequency.

Figures 2(a) and 2(b) show the measured intensity ratios I_{HH}^L/I_{HV}^T and the theoretically predicted graph given by (4) for both signs of P_{44} and both materials. The sign of P_{44} for amorphous material is also negative for static strain or for low frequencies [3].

In order to calculate a theoretical curve given by (4) we used for quartz the absolute values of the Pockels coefficients ($P_{12} = 0.27$, $P_{44} = \pm 0.0718$) [7]; the sound velocities are $c_L = 5920 \text{ m s}^{-1}$, $c_T = 3790 \text{ m s}^{-1}$ from [1].

For o-terphenyl at room temperature we found the ratio of the Pockels coefficients is $P_{44}/P_{12} = -0.094 \pm 7\%$ and the ratio between sound velocities $c_T/c_L = 0.38 \pm 3\%$; the latter result is in agreement with [9].

As one can see in figures 2(a) and 2(b) the Pockels coefficients P_{44} of o-terphenyl and quartz glass are both *negative*. The complete suppression of the LA line at the angle specified by (5) occurs at an angle $\theta \approx 85^\circ$ for o-terphenyl. For quartz this angle is $\theta \approx 70^\circ$. This result may prove useful in the spectroscopy of excitations of liquids and may give important information about the structure of liquids [3, 7].

The results of the present experiments are in agreement with qualitative theories which allow for $P_{44} < 0$ (see Mueller [3], Narasimhamurty [4] and Schroeder [7] for a discussion). However, they do not agree with the calculations suggested recently by Mazzacurati *et al* [12], who predicted a value of P_{11} bigger than that of P_{12} ; thus $P_{44} > 0$, for all glasses.

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Note: there is a printing error in their equation (7): the last symbol should be δ_i , instead of S_i .
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